1972-06

Preliminary investigation of collisional de-excitation rates for the (3)D-(3)P transition in helium I and (2)F-(2)D transition in helium II.

Harris, William Douglas.

PRELIMINARY INVESTIGATION OF COLLISIONAL DE-EXCITATION RATES FOR THE ^3D-^3P TRANSITION IN HELIUM I AND ^2F-^2D TRANSITION IN HELIUM II

William Douglas Harris

PRELIMINARY INVESTIGATION OF COLLISIONAL DE-EXCITATION RATES FOR THE ^3D-^3P TRANSITION IN HELIUM I AND ^2F-^2D TRANSITION IN HELIUM II

by

William Douglas Harris

Thesis Advisor: E. A. Milne

June 1972

Preliminary Investigation of Collisional De-Excitation Rates for the ^3D-^3P Transition in Helium I and ^2F-^2D Transition in Helium II

by

William Douglas Harris
Major, United States Army
B.S., Virginia Military Institute, 1962

Submitted in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE IN PHYSICS

from the

NAVAL POSTGRADUATE SCHOOL
June 1972

ABSTRACT

Helium gas was excited by 1.15 Mev protons from a Van de Graaff generator. Intensity of the radiation spectrum versus pressure data were recorded and plotted for the ^2F-^2D transition in He II (4686 Å). Through use of theoretical equations and experimental data, the collisional de-excitation rate coefficient, K, was determined to be $(5.03 \pm 0.21) \times 10^{-12}$ cm^3 sec^{-1}. Suggested experimental procedure is discussed for continuation of this investigation using electron excitation.

TABLE OF CONTENTS

LIST OF ILLUSTRATIONS

Figure

ACKNOWLEDGEMENTS

The author wishes to express his appreciation to Professor E. A. Milne for the continuing theoretical and technical guidance throughout this research. The author also would like to express his gratitude for the invaluable technical support provided by T. Maris and R. Moeller of the physics shop.

This research was supported in part by the Naval Ordnance Laboratory, White Oak, Maryland.

I. INTRODUCTION

Previous studies conducted at the United States Naval Postgraduate School have investigated the collisional de-excitation of argon and molecular nitrogen gas systems. In this work, a continuation of those studies, helium gas was bombarded with 1.15 Mev protons and the ^2P-^2D transition in He II (4686 Å) monitored for target chamber pressures ranging from 10 to 600 torr. From a plot of pressure divided by relative spectral line intensity, versus pressure, the collisional de-excitation rate coefficient was determined.

Original plans also called for the monitoring of the ^3D-^3P transition in He I (5876 Å) and the investigation of the "quenching" effect of nitrogen on both transitions. Due to deterioration of the proton accelerator system, a decision was made to convert the equipment to an electron accelerator. It is anticipated that future work utilizing electron bombardment will complete this investigation.

II. THEORY

A. GENERAL

The primary excitation reactions due to proton (H^+) impact upon helium in the target chamber are:

$$H^+ + He \longrightarrow He^* + H^+ \quad (1)$$

$$H^+ + He \longrightarrow He^+ + H \quad (2)$$

$$H^+ + He \longrightarrow (He^+)^* + H^+ + e^- \quad (3)$$

Equations (1) and (2) represent direct excitation and charge transfer, respectively, while equation (3) represents simultaneous excitation and ionization.

The primary de-excitation reactions occuring in helium are:

$$He^* \longrightarrow He + h\nu \quad (4)$$

$$(He^+)^* \longrightarrow He^+ + h\nu \quad (5)$$

$$(He^+)^* + He \longrightarrow He^+ + He + h\nu \quad (6)$$

$$He^* + He \longrightarrow 2\,He + h\nu \quad (7)$$

Equations (4) and (5) represent radiative de-excitation while equations (6) and (7) are collisional de-excitation.

The de-excitation of helium when in the presence of molecular nitrogen occurs through the following processes:

$$He^* + N_2 \longrightarrow He + N_2^* + h\nu \quad (8)$$

$$He^* + N_2 \longrightarrow He + N_2 + h\nu \quad (9)$$

$$(He^+)^* + N_2 \longrightarrow He^+ + N_2^* + h\nu \quad (10)$$

$$(He^+)^* + N_2 \longrightarrow He^+ + N_2 + h\nu \quad (11)$$

If a proton beam of current density J is incident on a target gas of density N atoms/cm^3, the rate of change in

population of excited state j of atomic species L with respect to time is given by:

$$\frac{dN_{Lj}}{dt} = \frac{J\sigma_{Ljg}N_{Lg}}{e} + \sum_{\substack{l \\ l>j}} \lambda_{Llj}N_{Ll} - \sum_{\substack{l \\ l<j}} \lambda_{Ljl}N_{Lj}$$

$$+ \sum_{M,i,l} K_{ilLM}N_{Ll}N_{Mi} - \sum_{M,i} K_{jiLM}N_{Lj}N_{Mi} \qquad (12)$$

where, L = particular atomic species under observation

M = any atomic (molecular) species.

l,i = energy states, with ground state denoted by g.

j = particular excited state under observation.

λ_{Llj} = transition probability for species L from state l to state j.

K_{jiLM} = collisional rate coefficient for de-excitation of state j in species L by collisions with atoms (molecules) of species M in state i.

σ_{Ljg} = cross section for excitation from ground state to state j of atomic species L.

The first term in equation (12) represents the rate of population of the excited state under observation due to direct excitation of the ground state. The second term is the population of the excited state j by radiative transitions from higher excited levels into state j. The third term reflects the de-population of state j by radiative transitions to lower excited states and the ground state. The fourth term is the excitation to state j due to collisions between atoms in state l of species L and atoms

(molecules) of type M in state i. The final term represents the de-population due to collisions between atoms in state j and atoms (molecules) of type M in the state i.

In the first term, the excitation cross section, σ_{Ljg}, is energy dependent. For any given experimental data run, the energy, current density, and target gas density were held constant. Hence, this term is constant. The assumption is made that the density of the target gas in state 1 is proportional to the partial pressure of the target gas. Therefore, the first term is proportional to P_L, the partial pressure of atomic species L. This assumption is justified due to validity of the ideal gas law in the pressure ranges of the study [Ref. 9]. The second term arises from transitions from higher excited states. This term is also assumed to be proportional to the partial pressure of the target gas, P_L. In the third term, the summation of transition probabilities to lower states is constant for a given spectral line. It is assumed that the population of excited state j, N_{Lj}, is proportional to the observed spectral line intensity thereby making this term proportional to $\lambda_{Lj}I$.

Applying the above assumptions at equilibrium conditions, equation (12) becomes:

$$\frac{dN_{Lj}}{dt} = 0 = aP_L - \lambda_{Lj}bI + \sum_{M,i} \frac{K_{ilLM}P_L N_{Mi}}{kT} - \sum_{M} \frac{K_{jiLM}bIP_M}{kT} \qquad (13)$$

where a and b are proportionality constants, k is Boltzmann's constant, and T is room temperature.

B. HELIUM AS TARGET GAS

When only helium is placed in the target chamber, equation (13) reduces to

$$0 = aP - \lambda_j bI - \frac{K_j bIP}{kT} \tag{14}$$

It is assumed that the probability of contributions from the third term on the right side of equation (13) due to collisions between excited helium atoms is small, and therefore can be neglected. Solving for the intensity in equation (14) yields

$$I = \frac{P}{A + BP} \tag{15}$$

where $A = \frac{\lambda_j b}{a}$

$$B = \frac{K_j b}{akT}$$

By plotting P/I versus P, a straight line should result with slope B and intercept A. Taking the ratio of B to A yields:

$$\frac{B}{A} = \frac{K_j}{\lambda_j kT} \tag{16}$$

Thus, the collisional de-excitation rate coefficient K_j can be determined if the transition probability for the spectral

line, λ_j, is known. The experimentally determined value of λ_j for the He I transition is 0.706×10^8 sec^{-1} [Ref. 7]. A literature search failed to uncover an experimental value for the He II transition. Using the relationship between Einstein coefficients for hydrogen and hydrogen-like species, the value was computed to be 0.548×10^8 sec^{-1} [Ref. 1]. The value of the temperature was taken to be 300 K.

C. HELIUM WITH MOLECULAR NITROGEN

For two gas systems where the "quenching" gas has excited state energies less than the excited atoms, N_{Lj}, equation (12) becomes:

$$0 = aP_L - \lambda_{Lj}bI - \frac{K_{jLM}bIP_M}{kT} - \frac{K_{jLL}bIP_L}{kT} \qquad (17)$$

Since helium is an inert gas, the excited state energy levels are high compared to those of molecular nitrogen [Ref. 3]. Therefore, there is de-excitation of helium due to collisions with other helium atoms and nitrogen molecules, but no excitation of the helium by collision with nitrogen molecules. Again, excitation to state j by collisions between helium atoms is assumed negligible. Equation (17) is rewritten as:

$$I = \frac{P_L}{A + BP_L + CP_M} \qquad (18)$$

where, P_L = partial pressure of helium

P_M = partial pressure of nitrogen

$$A = \frac{\lambda_{Lj} b}{a}$$

$$B = \frac{K_{jLL} b}{akT}$$

$$C = \frac{K_{jLM} b}{akT}$$

K_{jLM} = collisional de-excitation rate coefficient for atoms of type L in state j colliding with molecules of type M.

The constants A and B are determined by admitting helium alone into the target chamber and observing spectral intensities as helium pressure is incremented up to some final pressure. At this point, A and B are computed as in section B above. Holding the partial pressure of helium fixed, research grade molecular nitrogen is introduced in increments as the spectral intensity is again monitored. By plotting P_L/I versus P_M, a straight line of slope C and intercept $(A + BP_L)$ results. From the ratio

$$\frac{C}{A + BP_L} = \frac{K_{jLM}}{\lambda_{Lj} kT + K_{jLL} P_L} \qquad (19)$$

the rate coefficient for the quenching gas, K_{jLM}, is computed.

III. EXPERIMENTAL PROCEDURE

A block diagram of the experimental apparatus and associated electronic circuits is illustrated in Figure 1.

A proton beam produced by a 2 Mev Van de Graaff positive ion accelerator was accelerated down a drift tube and mass analyzed by a pair of bending magnets. The drift tube, at a pressure of approximately 2×10^{-6} torr, was physically separated from the target chamber by a one-half mil aluminum foil. The proton energy loss in passing through the foil is 0.25 ±0.05 Mev at the energies of this study [Ref. 4].

The target chamber consists of a pyrex glass tee with faraday cup attached for collecting the proton beam current. The beam current is fed to a switching circuit which can be set to monitor beam current by use of a Keithley Model 410 micro-microammeter. In this mode, the accelerator beam entering the target chamber is maximized at the desired energy. With the switching circuit in the alternate mode, the beam current is fed to a Keithley Model 610BR electrometer to record total charge collected at the faraday cup. The target chamber can be evacuated to 4×10^{-7} torr by a liquid nitrogen, trapped oil diffusion pump. This evacuation was accomplished approximately one hour prior to any experimental run.

The pyrex tee was attached to a manifold which provided gas inlet to the chamber and pressure monitoring. The

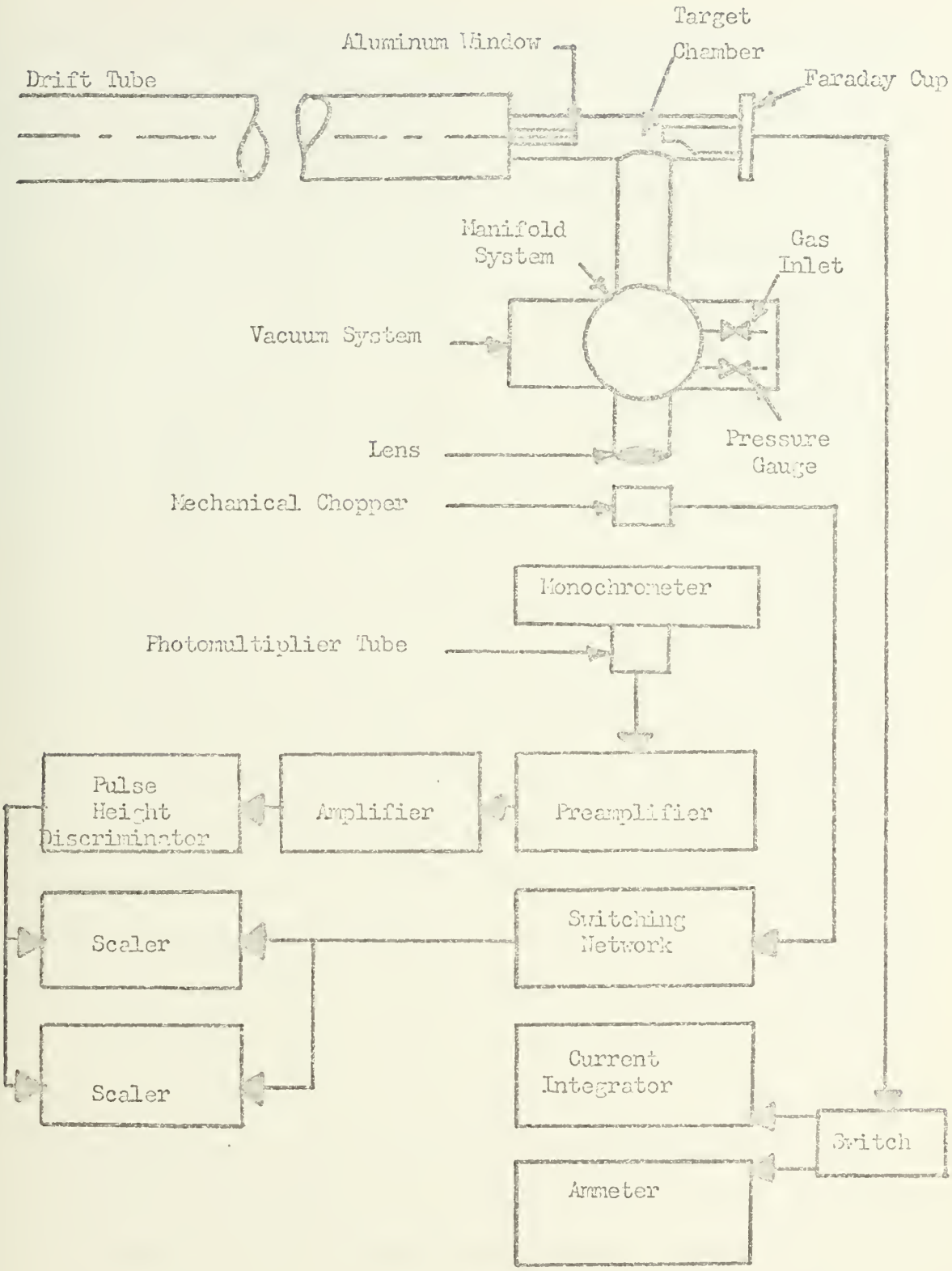

FIGURE 1. Block Diagram of Experimental Apparatus

pressure was measured by two Wallace and Tiernan pressure gauges. One gauge registered from 0 - 50 torr with an estimated error of ± 0.2 torr, and the other gauge registered from 0 - 800 torr with ± 3 torr error.

A quartz lens of 15 cm focal length was placed at an angle of 90° with respect to the incident proton beam. The reaction spectrum resulting from the first millimeter or so beyond the aluminum foil was focused by the lens, passed through a mechanical chopper operating at 85 Hz, and allowed to fall on the 250 micron entrance slit of a Jerral Ash monochromator. The resolving power of the monochromator was determined to be ± 7 Å about the central maximum. The selected spectral line was then passed to an Amperex XP 1110 photomultiplier tube. The spectral response of the tube was 40% and 100% maximum for the 5876 Å and 4686 Å lines, respectively. Prior to an experimental run, the alignment of the detection system and monochromator wavelength setting were maximized by sending the PM signal to a Princeton Applied Research Model HR-8 lock-in amplifier.

During each data run, the output of the PM tube was sent to an Ortec Model 101 preamplifier, a Canberra Model 810 amplifier, a Canberra Model 830 discriminator, and then to two Canberra Model 840 scalers. The scalers were gated by a signal from the mechanical chopper such that one scaler counted during periods when light passed through the chopper to the PM tube, and the other scaler counted during the "dark" periods of chopper rotation. A third scaler was

pulsed at 0.1 sec intervals by a Berkeley Model 903 double pulse generator to serve as a timer. The operation of the scalers and timer was controlled by a gating circuit used in conjunction with the electrometer. This permitted termination of the counting sequence when a specified charge from the proton beam was collected at the faraday cup.

The relative spectral line intensity (I) for each transition is determined by

$$I = \frac{N_c}{Q} \tag{20}$$

where N_c is the total corrected photon count and Q is the beam charge collected. The value of N_c is calculated from

$$N_c = N_1 - N_2 \tag{21}$$

where N_1 is the count of the scaler recording photon, background, and dark current pulses and N_2 is the count from the scaler recording background and dark current alone. N_1 is corrected for a system dead time of 1.7×10^{-6} sec.

The experimental procedure used in the study was to admit helium into the evacuated target chamber in increments from 10 to 600 torr. Pressure and intensity data were recorded for each increment with data reduction, calculations, and curve plotting accomplished by use of a Hewlett-Packard Model 9100A calculator.

IV. RESULTS

A. GENERAL

The collisional de-excitation rate coefficient for the ^2F-^2D transition in He II was determined to be (5.03 ±0.21) x 10^{-12} cm^3 sec^{-1}. An extensive literature search failed to reveal data concerning this de-excitation reaction so no data are listed for comparison. The assumptions made in the development of equation (15) seem valid in view of the correlation between experimental data and theoretical prediction. Other excitation effects, such as that by high energy electrons and X-rays were neglected in the development of equation (15).

A series of equipment malfunctions culminating with the separation of the proton accelerator tube in the Van de Graaff generator curtailed the investigation. Since several attempts to repair the tube using epoxy cements were futile, and an electron tube was available, the decision was made to convert the generator to electron acceleration. It is anticipated that electrons in the several hundred kilovolt range will provide sufficient excitation for reasonable resolution. The conversion is in progress at this time. Subsequent to completion of the conversion and modification of the laboratory area for protection against increased radiation hazards, the remaining portions of this study will be undertaken. A suggested procedure for this continuation in included in part B below.

B. SUGGESTED PROCEDURE USING ELECTRON EXCITATION

The primary de-excitation reactions for He I and He II are unaltered by use of electron excitation in lieu of protons. Therefore, the technique used in this study should serve equally well for investigation of the single gas He I transition. However, previous work has indicated that the helium spectra are very sensitive to nitrogen impurities. Essentially, minute quantities of nitrogen cause the helium spectrum to vanish. Hence, to determine the collisional de-excitation rate coefficient for molecular nitrogen on helium it is essential that an accurate method of introducing minute amounts of nitrogen be used. A suggested method involves the mounting of a gas tight syringe on the target chamber manifold. Any one of several commercial types could be used with appropriate piping. If the pressure and volume of the nitrogen gas in the syringe are known, then the partial pressure of nitrogen added to the target chamber by each injection can be computed from

$$P_C = \frac{V_S P_S}{V_C} \tag{22}$$

where P_C = partial pressure of nitrogen added

P_S = nitrogen pressure in syringe

V_S = volume of syringe

V_C = volume of chamber

The volume of the target chamber is 1794 cm^3. Using nitrogen at pressures of about 180 torr in a 0.5 ml syringe will permit chamber pressure increments of approximately 0.05 torr. It is estimated that increments of this order will be required for meaningful data. Using this arrangement, helium is first added to the chamber in increments up to a pressure of 100 torr or so. Pressure and intensity data are recorded for each point. This data will permit computation of the constants A and B in equation (18). Keeping the final helium pressure fixed, research grade nitrogen should now be added to the chamber in 0.05 torr increments. Pressure data are developed by computation and intensity is recorded for each point. Nitrogen is added in this manner until the helium intensity essentially falls to that of background. The procedure outlined in part C of section II will allow determination of the nitrogen rate coefficient.

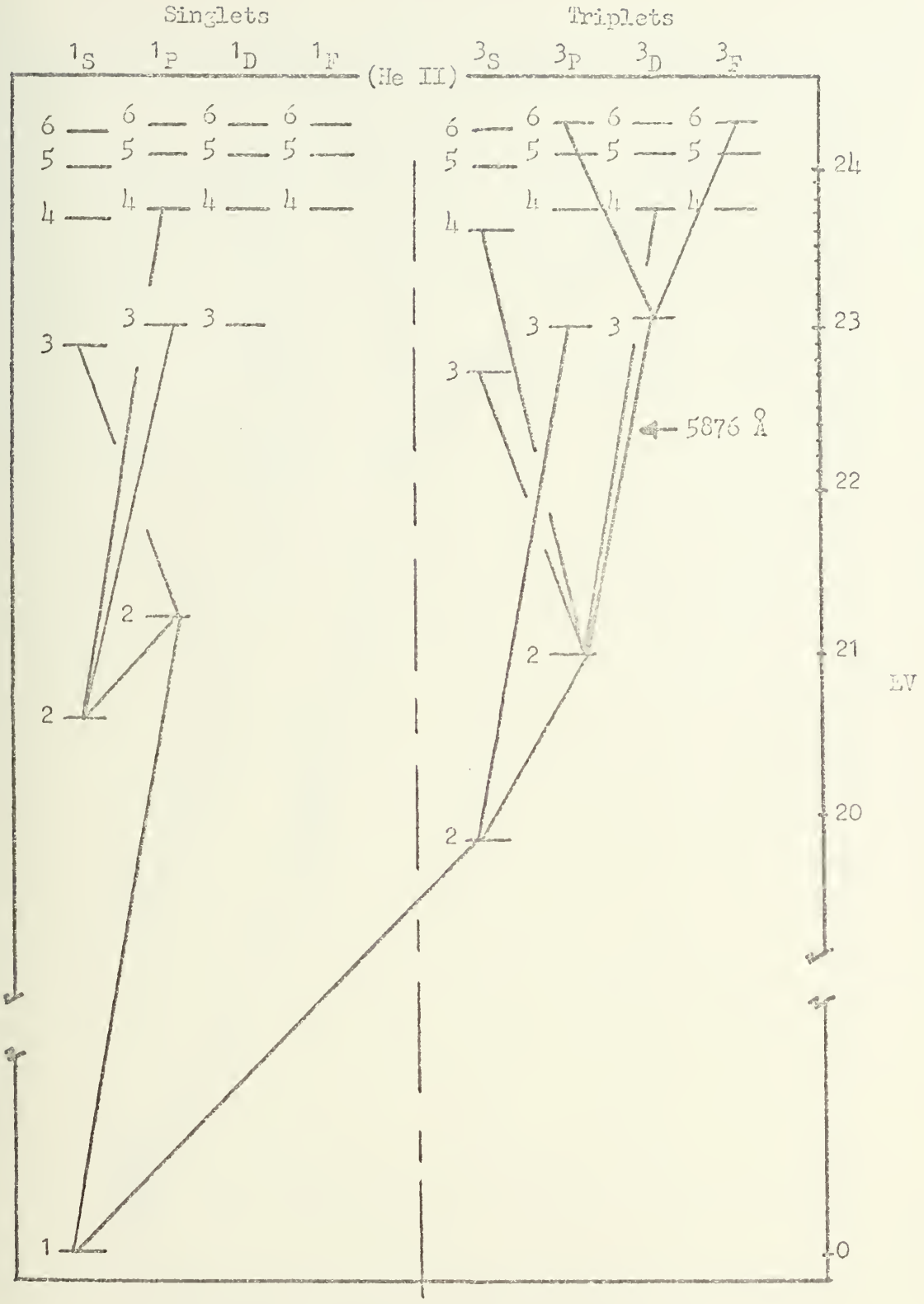

FIGURE 2. Energy Level Diagram; He I

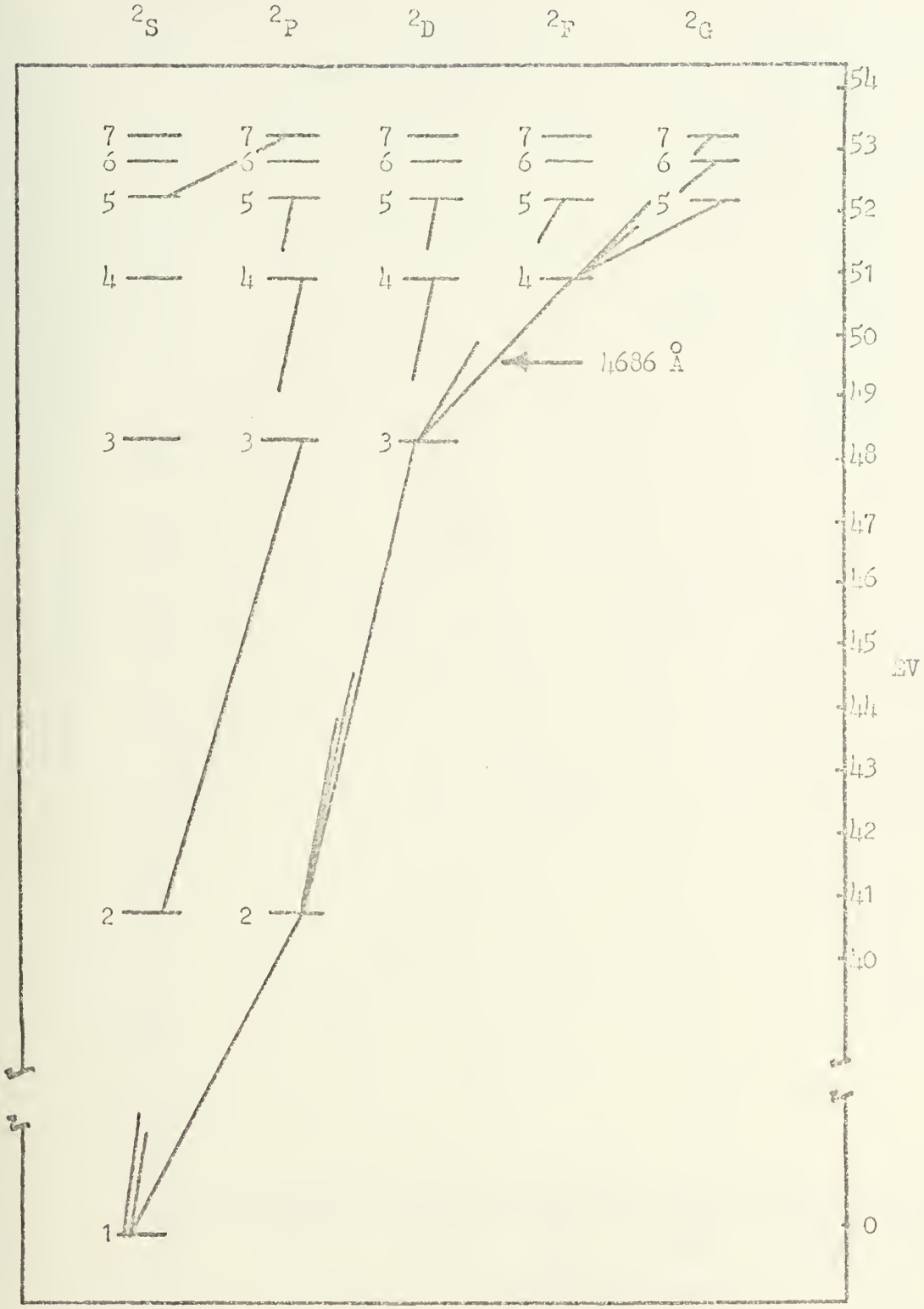

FIGURE 3. Energy Level Diagram; He II

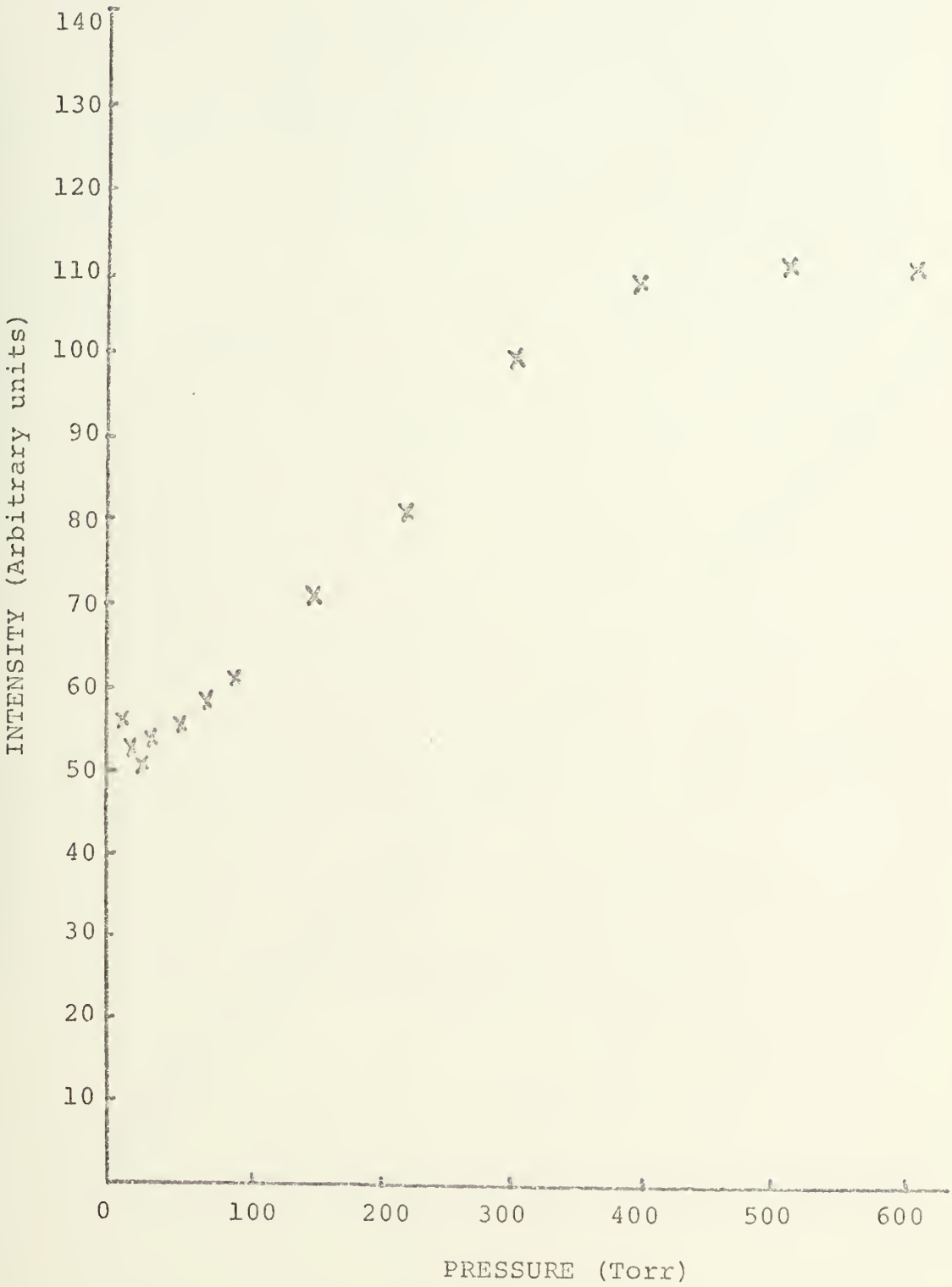

FIGURE 4. Intensity versus Pressure (4686 Å)

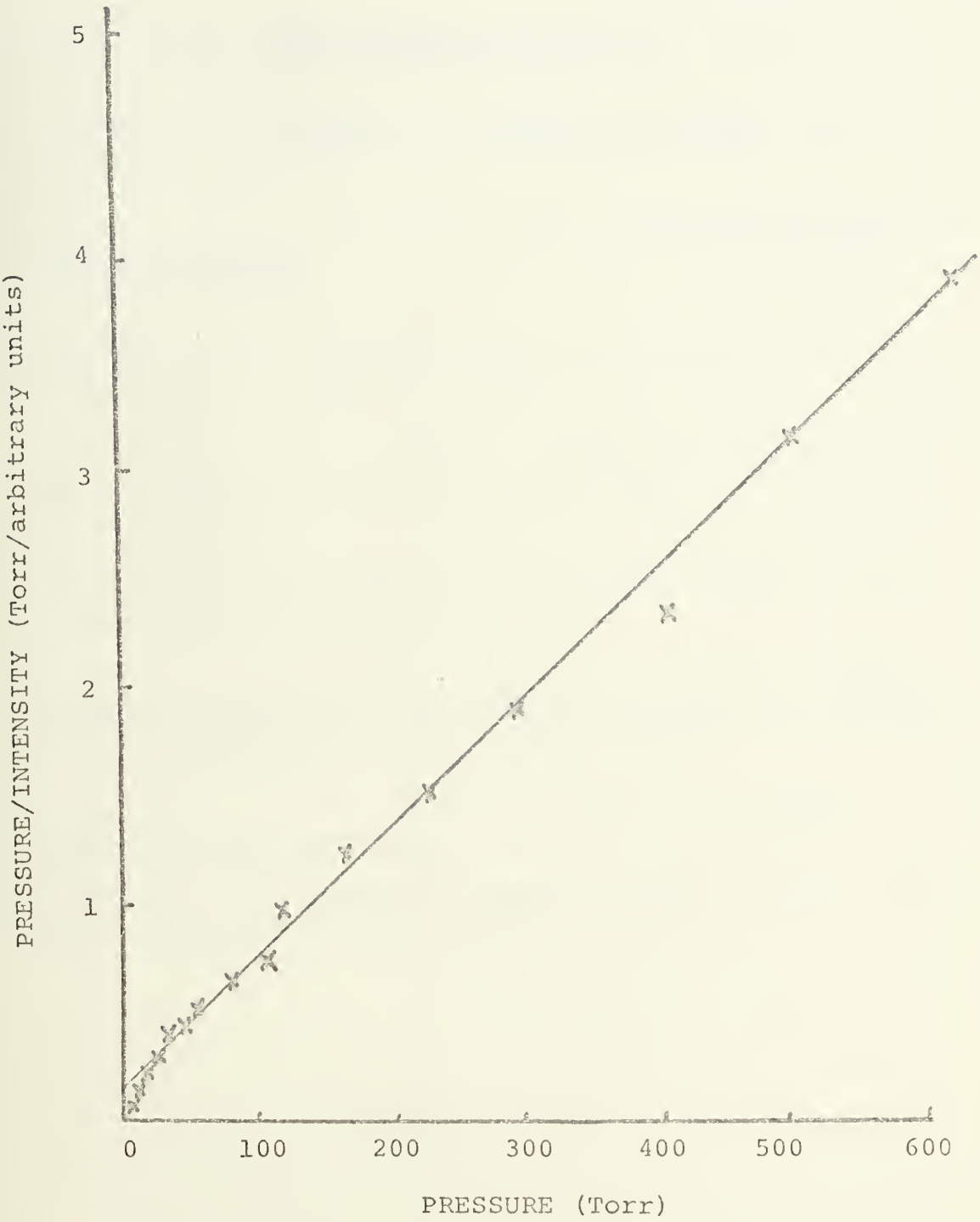

FIGURE 5. Pressure/Intensity versus Pressure (4686 Å)

BIBLIOGRAPHY

1. Allen, C. W., Astrophysical Quantities, The Athlone Press, University of London, 1955.

2. Gabriel, A. H. and D. W. O. Heddle, "Excitation Processes in Helium," Proceedings of the Royal Society, 258: 124-145, 1960.

3. Pearse, R. W. B. and A. G. Gaydon, The Identification of Molecular Spectra, 3rd ed., New York, John Wiley and Sons, Inc., 1963.

4. Smelley, A. R., "Reaction Rates for the Collisional De-Excitation of the $B^2\Sigma_u^+$ State of N_2 and the $C^3\Sigma_u$ States of N_2," Naval Postgraduate School Thesis, 1969.

5. Sternberg, Z. and P. Thomas, "Excitation Processes in Helium Induced by Impact of Deuterons and Protons," The Physical Review, 124: 810-813, 1961.

6. Tullington, B. J. Jr., "The Excitation of Helium by High Energy Proton Bombardment at Various Pressures," Naval Postgraduate School Thesis, 1968.

7. Wiese, W. L., M. W. Smith, and B. M. Glennon, Atomic Transition Probabilities, Hydrogen Through Neon, National Bureau of Standards, Department of Commerce, Washington, D. C., Government Printing Office, 1966.

8. Zaidel, A. N., V. K. Prokof'ev, and S. M. Raiskii, Tables of Spectral Lines, New York, Macmillan Co., 1961.

9. Zemansky, M. N., Heat and Thermodynamics, 4th ed., New York, McGraw-Hill Publishing Co., 1957.

INITIAL DISTRIBUTION LIST

		No. Copies
1.	Defense Documentation Center Cameron Station Alexandria, Virginia 22314	2
2.	Library, Code 0212 Naval Postgraduate School Monterey, California 93940	2
3.	Professor E. A. Milne, Code 61Mn Department of Physics Naval Postgraduate School Monterey, California 93940	1
4.	MAJ William D. Harris 20 Afton Parkway Portsmouth, Virginia 23700	1
5.	Mr. L. Rudlin Air/Ground Explosions Division Naval Ordnance Laboratory Silver Springs, Maryland 20910	1

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author)

Naval Postgraduate School
Monterey, California 93940

2a. REPORT SECURITY CLASSIFICATION

Unclassified

2b. GROUP

3. REPORT TITLE

Preliminary Investigation of Collisional De-Excitation Rates for the ^3D-^3P Transition in Helium I and ^2F-^2D Transition in Helium II

4. DESCRIPTIVE NOTES (Type of report and, inclusive dates)

Master's Thesis; June 1972

5. AUTHOR(S) (First name, middle initial, last name)

William Douglas Harris

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
June 1972	27	9

8a. CONTRACT OR GRANT NO.	9a. ORIGINATOR'S REPORT NUMBER(S)
b. PROJECT NO.	
c.	9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)
d.	

10. DISTRIBUTION STATEMENT

Approved for public release; distribution unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY
	Naval Postgraduate School Monterey, California 93940

13. ABSTRACT

Helium gas was excited by 1.15 Mev protons from a Van de Graaff generator. Intensity of the radiation spectrum versus pressure data were recorded and plotted for the ^2F-^2D transition in He II (4686 Å). Through use of theoretical equations and experimental data, the collisional de-excitation rate coefficient, K, was determined to be $(5.03 \pm 0.21) \times 10^{-12}$ cm^3 sec^{-1}. Suggested experimental procedure is discussed for continuation of this investigation using electron excitation.

DD FORM 1 NOV 65 1473 (PAGE 1)

S/N 0101-807-6811

Security Classification

A-31408

KEY WORDS	LINK A		LINK B		LINK C	
	ROLE	WT	ROLE	WT	ROLE	WT
Collisional De-Excitation						

Thesis
H2927 Harris
c.1

135223

Preliminary investigation of collisional de-excitation rates for the 3_D-3_P transition in helium I and 2_F-2_D transition in helium II.